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A μSR and neutron scattering study of $REGa_6$, where RE = Ce, Nd, Gd and Tb

Erik Lidström†, Roger Wäppling†, Ola Hartmann†, Mikael Ekström† and G M Kalvius‡

† Department of Physics, Uppsala University, Box 530, S-751 21 Uppsala, Sweden
† Physics Department, TU Munich, D 85747 Garching, Germany

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Abstract. The $REGa_6$ series presents varied magnetic ordering behaviour within compounds that are structurally identical except for slight differences in lattice parameters. The compounds are quasi-two-dimensional and order at temperatures between 1.7 and 20 K. The two magnetic structures of $TbGa_6$ have been refined and the results compared to a previously unpublished magnetic neutron diffraction study of most of the series. All four compounds in this study have been examined using zero-field and longitudinal-field μSR. In $CeGa_6$, $NdGa_6$ and $TbGa_6$ a systematic build-up of magnetic correlations occurs as the magnetic ordering temperature is approached, while in the Gd compound no increase of magnetic correlations is observed within the experimental resolution of 0.1 K. In $NdGa_6$, a linear increase is observed in a log–log diagram of the muon depolarization rate as the temperature is decreased from 30 times the ordering temperature towards T_N. In three of the compounds ($NdGa_6$, $GdGa_6$ and $TbGa_6$), two distinct muon sites are observed. The exponents for the damping as a function of the reduced temperature are found to be dependent on the muon site, especially in $TbGa_6$ where the signals from the two different sites can be best separated.

1. Introduction

The series $REGa_6$, where RE is a rare-earth metal, is a quite recently discovered group of compounds [1] that crystallize in the tetragonal $PuGa_6$-type structure with space group $P4/nbm$ [2]. As shown in figure 1, layers of rare-earth atoms are stacked perpendicular to the c-axis with four layers of gallium between each pair of rare-earth layers. The distance between rare-earth atoms in the c-plane is about 4.2 Å while the distance between the layers is about 7.6 Å. Interactions between the rare-earth atoms in the plane can, therefore, be expected to be much more important than those between different planes; hence quasi-two-dimensional properties might be anticipated. Further, magnetic correlations can be expected to be important within the planes far above the ordering temperature. The understanding of magnetical fluctuation phenomena is far from complete and it was anticipated that the study of the present series of materials might give rise to interesting observations. Because of its sensitivity to magnetic fluctuations, as discussed below, μSR is an obvious experimental tool.

Most of the members of the series order antiferromagnetically [3, 4]. YGa_6 superconducts at ambient pressure whilst $TbGa_6$ and $DyGa_6$ become superconducting only at pressures of about 10 kbar with T_c being about 6 K [5]. A summary of the magnetic properties of the materials included in the present study is given in table 1.

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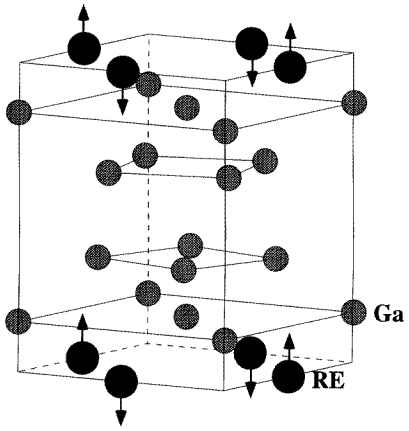

Figure 1. The crystal structure of the $REGa_6$ compounds. The arrows on the rare-earth atoms show the magnetic ordering of $TbGa_6$ in the commensurate phase.

Table 1. A summary of the magnetic properties of the $REGa_6$ compounds investigated in the present study: (a) the present study; (b) Sakurai *et al*; (c) Jerjini; (d) μ_{eff}^{obs} is the effective moment deduced from magnetization measurements by Sakurai *et al*; (e) deduced from the crystal-field levels determined by Jerjini.

Compound	Ordering temperature (K)	Type of ordering	$\boldsymbol{k}$-vector	Moment direction	μ_{eff}^{obs} [d] (μ_B)	Saturation moment (μ_B)
$CeGa_6$	1.7(1)[a]	—	—	*a*–*b*-plane[c]	2.37	0.4[e]
$NdGa_6$	10.4[b]	A.F.	(0, 0, 1/2)	*c*-axis	3.31	2.45(15)[c]
$GdGa_6$	12.50(5)[a]	A.F.	—	—	—	—
$TbGa_6$	20.2(3)[a]	A.F.	(k, k, 0)	*c*-axis	9.80	
	14.5(2)[c]	A.F.	(0, 0, 0)	*c*-axis		7.98(2)[a]

Given the layered structure of these materials, it would be desirable to perform experiments on single crystals. Unfortunately, the compounds are formed in peritectic reactions, making it very difficult to grow single crystals. Attempts by Jerjini [4] met with limited success. Polycrystalline samples were used in the present μSR and neutron scattering study.

2. Experimental details

2.1. Sample preparation and characterization

The samples were prepared by arc-melting the constituent elements on a water-cooled copper hearth in an argon atmosphere. They were then heat treated under vacuum at about 50 K below the peritectic temperature for one to two days. X-ray diffraction films obtained in a Hägg–Guinier camera showed that the samples were of good quality with the maximum level of impurity phase being 3% by weight.

2.2. Neutron scattering

Neutron diffraction measurements were made on $TbGa_6$ using the POLARIS medium-resolution time-of-flight diffractometer at the ISIS spallation source, UK, and the fixed-wavelength powder diffractometer (NPD) at the R2 research reactor, Studsvik, Sweden.

NPD is equipped with a double-crystal copper monochromator giving a wavelength of 1.470(1) Å. At both diffractometers, the sample was contained in a cylindrical vanadium canister. At ISIS the sample was mounted in an ILL 'Orange' liquid helium cryostat, while at Studsvik a closed-cycle refrigerator (CCR) was used.

It was known from magnetic susceptibility data that there are two antiferromagnetic transitions in $TbGa_6$, one at about 18 K and another between 14 and 15 K [3]. The ISIS data were collected at 22, 17 and 4.5 K to obtain data for each magnetic region. The NPD data were obtained at 22, 18, 16, 15, 14 and 11 K to study the development of the magnetic structure that had been determined from the ISIS results. Measurements were performed between $\sin\theta/\lambda = 0.047$ and 0.22 $Å^{-1}$. The data from the POLARIS low-angle A-bank (centred at 20° in 2θ) and the back-scattering C-bank (centred at 145° in 2θ) were focused in time of flight, normalized to the incident beam monitor and corrected for instrumental background and detector efficiency. Rietveld refinements were made using the programs TF14LS for the nuclear-only refinements of the paramagnetic phases and TF104M for the antiferromagnetic phases. These programs form part of the Cambridge Crystallographic Library of subroutines and programs [6]. The NPD data were analysed using the FULLPROF program [7]. Nuclear scattering lengths were taken from [8]. The crystal structure was first refined in the paramagnetic phase using the C-bank data covering a range in $\sin\theta/\lambda$ from 0.16 to 2.5 $Å^{-1}$. The atomic coordinates were then fixed in the subsequent refinements of the A-bank forward-scattering data, which covered a range in $\sin\theta/\lambda$ from 0.06 to 1.0 $Å^{-1}$. Magnetic scattering decreases rapidly with increasing $\sin\theta/\lambda$, so its contribution to the diffraction pattern in the A-bank detectors is larger than it is in the C-bank. As a consequence, although the A-bank has poorer resolution, the data it provides are usually crucial to the accuracy with which the magnetic structure can be determined. The magnetic form factor for the Tb^{3+} ion was taken from [9].

2.3. μSR spectroscopy

Measurements of all samples were made at the ISIS spallation source at DRAL, UK, whilst $TbGa_6$ and $NdGa_6$ were also studied, at the TRIUMF facility in Canada and at PSI in Switzerland, respectively. The samples were studied using both zero-field and longitudinal-field μSR spectroscopy. Weak-transverse-field (∼2 mT) data were taken at selected temperatures to fix the initial asymmetry.

At ISIS, the data were collected from 10 to 300 K in a CCR, and between 40 mK and 4 K in a dilution cryostat. At TRIUMF, a helium-flow cryostat was used for measurements between 3.5 and 300 K. At PSI, data were obtained between 10 and 40 K in a CCR. The ISIS surface muon beam is pulsed (with a pulse width of approximately 70 ns) and has a negligible background. It is therefore best suited for studies of materials where the muon depolarization is fairly slow (i.e., depolarization rates between 3 and 0.01 μs^{-1}). In all of the compounds in the present study, there were temperature regions where the depolarization rate was outside the accessible region of the instrument used.

The zero-field and longitudinal-field μSR technique is described in detail elsewhere, e.g. in [10, 11], and only a short description is given here. The muons enter the sample with their polarization parallel to the beam direction, chosen as the z-direction. As a muon decays, it emits a positron preferentially in the direction of the spin at the time of decay. The polarization of the muons is measured as the difference between the numbers of positrons detected in the forward and backward detectors, i.e.

$$a_0 G(t)_z = \frac{N(t)_F - \alpha N(t)_B}{N(t)_F + \alpha N(t)_B}$$

where a_0 is the initial polarization (asymmetry) and $G(t)_z$ is the depolarization function containing the experimental information. $N(t)_F$ and $N(t)_B$ are the numbers of detected positrons in the forward and backward detectors and α is a factor correcting for differences in detector efficiency, geometry, etc.

Implanted muons thermalize very quickly and usually come to rest on interstitial sites in metals. Depending on the temperature and the material, they can remain immobile or diffuse during the time of observation (typically at least 10 μs). Depolarization of muons is caused either by static or by fluctuating magnetic fields. In zero-field μSR for the case of static, randomly ordered, concentrated magnetic moments, the distribution of the internal field is Gaussian and the depolarization function will have the static Kubo–Toyabe shape [10]. Its prominent feature is a Gaussian ($\sim e^{-\Delta^2 t^2}$) decay of polarization at short time and a recovery polarization equal to one third of the initial asymmetry, a_0, at long times. The factor Δ is the static width, that is, a measure of the breadth of the distribution of the field at the muon site which causes dephasing of the muon signal. This situation is mainly realized if the local field has nuclear dipole moments as a source.

In the case where the muons sense rapidly fluctuating fields, the Kubo–Toyabe function approaches an exponential decay (Lorentzian shape), $e^{-\lambda t}$. This can be the case if the muons are static and there are rapidly fluctuating moments (spin dynamics) or if the muons diffuse rapidly and magnetic ions are concentrated in the lattice (muon dynamics). In both cases, $\lambda = \gamma_\mu^2 \langle B^2 \rangle \tau_c$, where the gyromagnetic ratio for the muon is given by $\gamma_\mu/(2\pi)$ = 135.54 MHz T^{-1} and $\langle B^2 \rangle$ is the average of the square of the magnetic field at the muon site (provided that the average field is zero, which holds for certain in a paramagnet in low applied field). τ_c is the correlation time for the fluctuations, in the case of static muons, or, if the muons are mobile, the diffusion correlation time. Muon diffusion can usually be disregarded in the presence of fluctuating moments, since the correlation time for diffusion is much larger than that of the magnetic moments [12, 13].

In a magnetically ordered material the muons will precess in the local field at the muon site. In the case of antiferromagnets, the field may cancel at the muon site because of symmetry. In the magnetically ordered state in a randomly oriented powder, with a Gaussian spread of magnetic fields on the muon sites, one obtains two interconnected signals:

$$\frac{1}{3} a_0 e^{-\lambda t} \tag{1}$$

and

$$\frac{2}{3} a_0 e^{-\lambda t} e^{-(\Delta t)^2} \sin(\omega t + \phi) \tag{2}$$

where a_0 is the modulus of the initial polarization, λ the Lorentzian depolarization, ω is the angular frequency, directly proportional to the mean field at the muon site, and ϕ is the initial phase. Provided that the dephasing of the signal, expressed as $e^{-(\Delta t)^2}$, is caused by nuclear moments, the values of Δ should be close above and below the magnetic ordering temperature. The non-rotating part stems from the average third of the local fields that are parallel to the z-axis. If the dephasing is very rapid one might not observe the second signal. The non-rotating first part often decays very slowly, and if the second part is not resolved, the magnetic transition might be observed as a shift in the base-line.

2.4. μSR and magnetic phase transitions

Rapid magnetic fluctuations are usually present far above the magnetic ordering temperature. They slow down on approaching the phase transition, especially very close to the transition

temperature (the critical region) in accord with the formalism of spin correlations. Within the critical region our understanding of the detailed nature of spin fluctuations is incomplete. Present theoretical treatments deal with ideal cases of pure Ising behaviour or the isotropic Heisenberg interactions [14, 15, 16]. In addition it has been shown that dipolar interactions between the localized spins play a significant role [17, 18]. The problem of anisotropy in the exchange interactions, surely important in rare-earth materials, remains a challenge to date.

μSR is a very useful tool for the study of magnetic fluctuations and critical phenomena. Because of its great sensitivity to small moments and its wide time window it is often possible to study magnetic fluctuations at temperatures many times the ordering temperature, as well as very close to it [19]. Another advantage over many other techniques is that μSR spectroscopy can be performed at true zero field so as to minimize disturbance to the system under study. A fact that can be of great use in the study of single crystals is that the muon is not sensitive to the slowing down of spin fluctuations along its spin direction.

The technique also has some disadvantages. The magnetic field at the muon site is the vector sum of the isotropic contact and anisotropic dipole fields. The contact field can be determined through muonic Knight shift measurements [11, 20]. The very presence of the muon makes the contact field very difficult to calculate, partly because the muon causes lattice expansion locally, and partly because in metals the muon is screened by a cloud of conduction electrons. The dipole field is easily calculated, but its evolution close to a magnetic phase transition is difficult to ascertain, as discussed by Nishiyama *et al* [21] and more recently by Lovesey *et al* [22].

To extract quantitative information on the behaviour of magnetic fluctuations from muon data, one may need to calculate the dipole tensor for all $\boldsymbol{k}$-vectors. This is illustrated in table 4 of Lovesey *et al* [22] where it is shown that both the absolute value and the temperature dependence of the depolarization rate are different for different assumed muon sites in $RbMnF_3$. These authors have also calculated the critical depolarization behaviour, approximating the dipolar contributions with the dipolar tensor for the ordering vector. In the case where the muon sits in a field-free position in the ordered state, this approximation clearly cannot be made. It would mean that the muon depolarization is zero at the ordering temperature, contrary to experimental observations. At muon sites where the field is zero or very low in the ordered state it is therefore reasonable to assume that, to quantitatively extract the critical magnetic behaviour of the sample from the muon data, one has to take into account contributions from dipolar tensors other than that of the ordering vector.

Table 2. Results of the refinement of the C-bank data for $TbGa_6$ at 25 K. The coordinates are given according to Choice two in *The International Tables for Crystallography* with the origin displaced by $(\frac{1}{4}, \frac{1}{4}, 0)$ from that of Choice one. Choice one is used in all other references to crystallographic information in the paper.

a (Å)	5.912 56(2)
c (Å)	7.543 61(5)
Ga(4g), z	0.161 68(15)
Ga(8m), x	0.067 56(9)
Ga(8m), z	0.352 33(12)
R_{wp} (%)	1.82
$R_{w.int.}$ (%)	2.84

The correlation time, τ_c, for magnetic fluctuations, which contributes linearly to the depolarization rate λ, usually increases close to the magnetic transition and the size of

locally ordered clusters grows. In the present study, for a site such as (0, 0, $\frac{1}{2}$) in $NdGa_6$, the effective magnetic field is zero below T_N because of opposing contributions from different magnetic sublattices in the particular magnetic structure. As the order in the paramagnetic clusters reflects the situation in the ordered state, this means that as the correlations increase, the net field decreases. This effect therefore works in the opposite direction compared to the effect of the increased correlation time.

The question of extracting information on magnetic fluctuations from muon data warrants further theoretical study.

3. Results and discussion

3.1. Crystallographic and magnetic structures

The present study was commenced without knowledge of the unpublished neutron diffraction study on the series by Jerjini [4]. We report our own neutron scattering results for $TbGa_6$, comparing them with Jerjini's results, and also report relevant information on the magnetic structure of the rest of the series obtained by him.

The magnetic structures for $GdGa_6$ and $CeGa_6$ have not been determined, as far as we are aware. Because of the high gadolinium neutron absorption cross section, it is not possible to study $GdGa_6$ with neutron diffraction without resorting to isotope substitution. $CeGa_6$ poses the problem of how to make a magnetic structure determination when the ordering temperature is below the range accessible to a normal helium-flow cryostat.

The results from nuclear refinement of the ISIS C-bank data for $TbGa_6$ are summarized in table 2. The crystal structure is displayed in figure 1.

The periodically ordered magnetic moments in a crystal can be decomposed in a Fourier sum over the $\boldsymbol{k}$-vectors:

$$\boldsymbol{m}(\boldsymbol{r}_l) = \sum_{\boldsymbol{k}} \boldsymbol{m}_k \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_l}. \tag{3}$$

The amplitude of the oscillation of the moments is A_k, defined as

$$\boldsymbol{m}_k = \frac{A_k}{2}\mathrm{e}^{\mathrm{i}\phi_k}\boldsymbol{u}_k \tag{4}$$

where $\boldsymbol{u}_k$ is a unit vector parallel to the Fourier component.

At 4.5 K peaks appear at the forbidden nuclear reflections (1 0 0) and (1 0 1), showing that the magnetic propagation vector is $\boldsymbol{k} = (0, 0, 0)$; see figure 2. Since $TbGa_6$ is an antiferromagnet, this means that the two terbium moments in each unit cell must be antiferromagnetically coupled to each other. The amplitude relationships show that the moments on the two terbium atoms in the unit cell are parallel and antiparallel to the c-axis; see figure 1. We refined a moment of 7.98(2) μ_B at 4.5 K in reasonable agreement with Jerjini [4] who found 8.9(5) μ_B at 4 K. The free Tb^{3+} moment is 9 μ_B.

Magnetic reflections corresponding to a propagation vector $\boldsymbol{k} = (0.08, 0.08, 0)$ were found in the 18 and 16 K NPD measurements and in the 17 K data from ISIS. The compound is thus incommensurately ordered. The structure has been refined assuming single-$\boldsymbol{k}$ domains and we find A_k, the amplitude of the modulation of the magnetic structure, to be 6.2(1) μ_B at 16 K and 3.6(1) μ_B at 18 K. At 17 K we obtained 5.5(1) μ_B from the ISIS data. These results compare well with Jerjini's 6.3(3) μ_B at 16 K.

At 15 K, the NPD data showed peaks originating from both of the magnetic structures. This could be due to sample inhomogeneity, a temperature gradient in the sample, or to the actual coexistence of the two phases. Jerjini measured a large number of temperatures

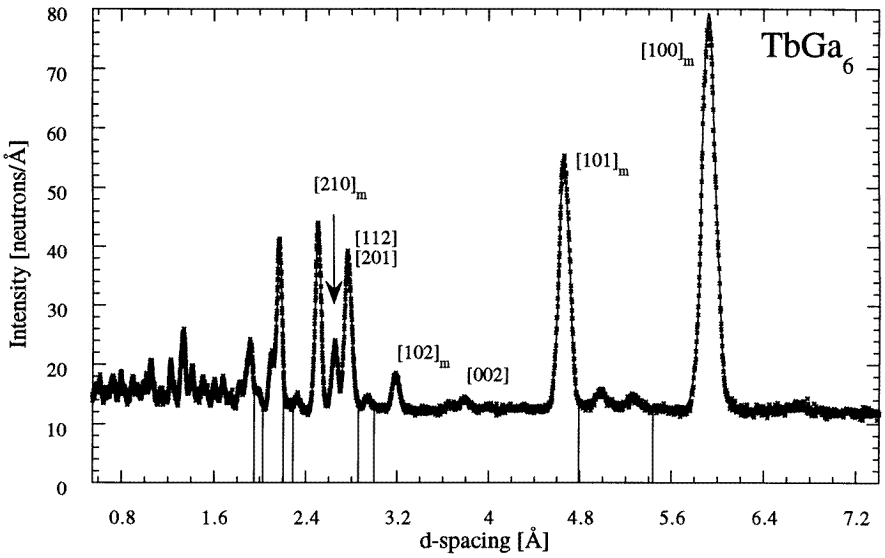

Figure 2. The Rietveld refinement of the 4.5 K $TbGa_6$ data obtained at ISIS. The vertical lines indicate regions excluded in the refinement.

Table 3. Results of the refinements of the magnetic structures for $TbGa_6$.

Facility	ISIS		Studsvik			
T (K)	4.5	17	11	14	16	18
$(k, k, 0)$	0	0.083	0	0	0.0797(3)	0.0820(4)
Moment (μ_B)	7.98(2)	5.50(4)	4.74(5)	4.31(4)	6.20(6)	3.64(6)
R_{wp} (%)	3.21	1.94	7.86	6.18	5.99	5.26
R_{Bragg} (%)	3.91	4.85	6.19	5.21	5.16	5.48
$R_{magnetic}$ (%)			7.91	4.74	7.22	17.3

close to the transition but did not report such coexistence; thus the second explanation is the most likely. The transition temperature found in [4] was 14.5(2) K. The results of the magnetic refinements are summarized in table 3.

There were also two additional, but weak, peaks appearing in the ISIS 4.5 K data. One of these is also visible as a shoulder on one of the magnetic peaks in the 17 K data. The reflections were too weak to be observed at the NPD and were not observed by Jerjini. These two peaks can be indexed as $(1\ \frac{1}{2}\ 0)$ and $(1\ \frac{1}{2}\ \frac{1}{2})$ indicating that a doubling of the unit cell occurs in the a- and c-directions. No $(0\ \frac{1}{2}\ 0)$ or $(0\ \frac{1}{2}\ \frac{1}{2})$ peaks were observed and the hypothetical $(0\ 0\ \frac{1}{2})$ peak was outside the range of the diffractometer at the time of the measurement. It is not possible to determine whether these peaks are of magnetic or nuclear origin. No lowering of the symmetry was observed in the high-resolution C-bank data. Since the extra peaks appear in the magnetic temperature regions, they could be of magnetostrictive origin. Judging by the good fits to the data, when these two peaks were excluded, the change in atomic coordinates, or magnetic moment distribution, must be small.

$NdGa_6$ orders antiferromagnetically below 10.2(2) K [4]. The magnetic propagation vector is (0 0 $\frac{1}{2}$), i.e., the magnetic unit cell is twice the size of the nuclear one along the *c*-axis. The magnetic ordering consists of ferromagnetically ordered sheets stacked along the *c*-axis with the spins alternatively up or down. The ordered moment at 3 K was found by refinement to be 2.45(15) μ_B as compared to the measured free-Nd^{3+} moment of 3.5 μ_B.

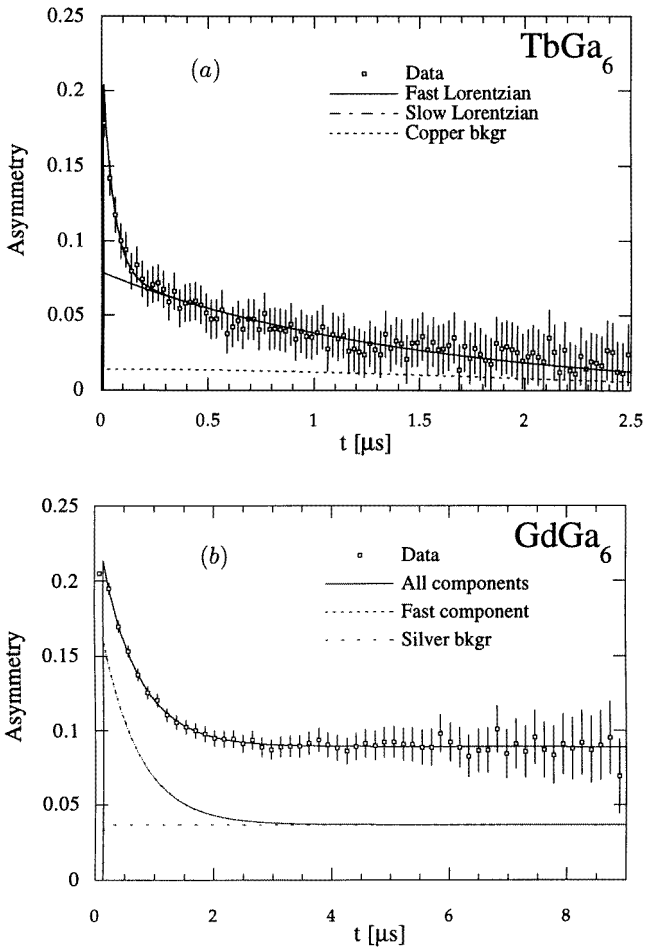

Figure 3. (a) The muon spectrum of $TbGa_6$ obtained in zero field at 45.3 K at TRIUMF. The spectrum has been fitted with two Lorentzian depolarization functions as well as a Gaussian one accounting for muons stopping in the copper sample holder. (b) Lorentzian depolarization in $GdGa_6$ obtained in a 20 mT longitudinal field at 17 K at ISIS. One component of the muon spectrum is completely decoupled by the field while the other is not affected.

For completeness, we summarize Jerjini's other magnetic structure results [4]. $HoGa_6$ and $DyGa_6$ order with the same magnetic structure as the incommensurate structure found in $TbGa_6$, except that there is a squaring up of the modulation at low temperatures, shown by reflections compatible with third-order harmonics of the $\boldsymbol{k}$-vector. $PrGa_6$ orders with the same structure as $NdGa_6$.

3.2. μSR results

3.2.1. Muon sites. As discussed in more detail below, two different muon signals were found in the neodymium, gadolinium and terbium compounds, whilst only one is seen in $CeGa_6$. The straightforward explanation is that the muon occupies two different sites in the former compounds. Two muon spectra illustrating this fact are shown in figure 3. It is deduced that the site giving the lowest exponential depolarization rate in $NdGa_6$, whose magnetic structure is known, must be a site where the net field in the ordered state is zero. This is because the depolarization of this component is easily decoupled in a longitudinal field—see figure 3. For the $(0, 0, \frac{1}{2})$ site in figure 1, the fields do cancel, and a calculation of the Gaussian depolarization caused by the randomly oriented gallium nuclear dipoles gives $\Delta = 0.22\ \mu s^{-1}$. The refined structural parameters for $TbGa_6$ were used in the calculation. The μSR fits give values that are in good agreement, $\Delta = 0.18(2)\ \mu s^{-1}$ in the case of $NdGa_6$ and $\Delta = 0.20(1)\ \mu s^{-1}$ for $CeGa_6$. For $GdGa_6$ it was difficult to obtain an accurate value for Δ from the fit, but it is possible to use the above-mentioned calculated value as a fixed parameter.

In the case of $TbGa_6$, the value of the Lorentzian depolarization rate of the slow component, coming from the electronic moments, was too high at 300 K to allow Δ to be determined. These more slowly depolarizing muons also sit in a field-free position in the ordered state, that is likely to be the same as in the other compounds given the similarities in the structural parameters. Other field-free positions, such as $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, give values for Δ that are far from those measured.

Since the depolarization rates of the fast components are much larger, the muons must be located much closer to a rare-earth atom, possibly in the rare-earth plane. It is not possible from the present study to identify these sites, but a likely candidate is midway between two rare-earth atoms in the rare-earth plane.

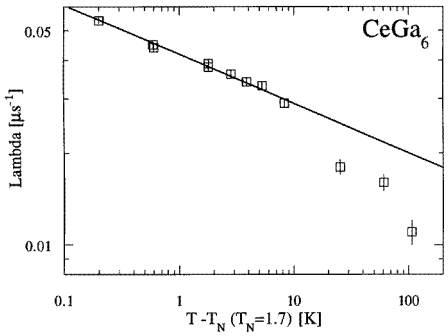

Figure 4. A double-logarithmic plot of the muon depolarization rate in $CeGa_6$ as a function of the reduced temperature.

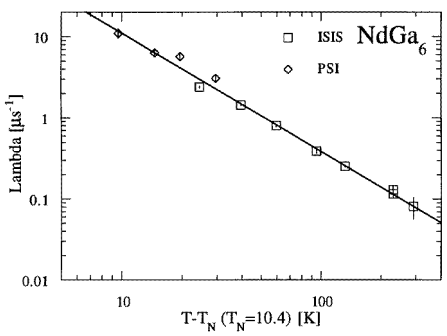

Figure 5. A double-logarithmic plot of the depolarization rate of the fast-muon signal in $NdGa_6$ versus temperature. The small difference between the two sets of data might be due to differences in the temperature calibrations.

3.3. $CeGa_6$

The muon depolarization observed in $CeGa_6$ was much weaker than the one in the other three compounds, so it was impossible to determine whether there are one or two components present. The weak depolarization is partly explained by a low Ce^{3+} moment. Jerjini found a

moment of about 0.4 μ_B for the ground-state crystal-field level. The Gaussian contribution to the depolarization in zero field is, however, compatible with all of the muons stopping in the $(0, 0, \frac{1}{2})$ site. The data obtained in a 10 mT longitudinal field were fitted with a Lorentzian function and an undamped background. The data had a fast initial component between 1.5 and 1.8 K, indicating the onset of magnetic order. Since the sample was a powder, absolute temperature homogeneity was not guaranteed, and although it was not possible to tell whether the temperature interval over which the ordering took place arose from sample inhomogeneity, the good quality of the x-ray diffractograms makes a temperature gradient in the sample the likely explanation. The depolarization rate is displayed in figure 4 as a function of $T - T_N$ with $T_N = 1.7$ K. The depolarization rate increases, starting from 10 K above the ordering temperature, with a power law $\lambda \sim (T - T_N)^{-0.46(5)}$ as the temperature decreases. Below the transition temperature, the amplitude was reduced to about one third of the value obtained in the paramagnetic region.

Above 12 K, the increase in depolarization with decreasing temperature is much steeper, and it therefore seems as if the strength of the magnetic correlations changes. Jerjini [4] also measured the crystalline electric field using inelastic neutron scattering. He reports two excited levels at 43 and 93 K. It is therefore possible that the change in the exponent comes about when only the lowest level is populated, but the reason for the change could also be the development of magnetic clusters (see the discussion on $TbGa_6$ below).

Jerjini stated that the susceptibility data indicate ferromagnetic ordering. From the muon data we cannot confirm whether this is the case. The spectra obtained in a weak transverse field of 2.0 mT give a weak negative Knight shift. A negative Knight shift can be observed in both antiferromagnets and ferromagnets if the contact field at the muon site is negative.

The specific heat measurements by Jerjini [4] show three peaks, indicating that there are three magnetic transitions. No abrupt changes in the amplitude or depolarization rates were observed below T_N in the present data to substantiate this, but because of the limited time resolution at ISIS, and the possible temperature uncertainty, more than one magnetic transition temperature cannot be completely ruled out. If all magnetic structures below T_N are ferro- or ferrimagnetic, the contact field will always be present. In the case where one or more of the possible structures is antiferromagnetic, it is possible that the muon could sit in a field-free position in this magnetic phase. The full asymmetry might then be observed.

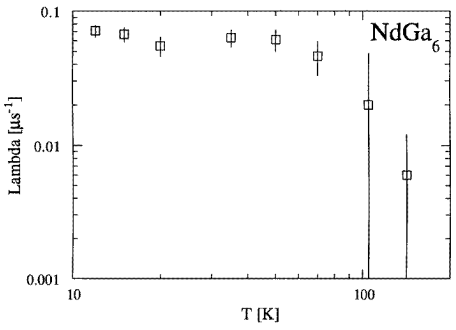

Figure 6. The double-logarithmic plot of the depolarization rate of the slow-muon signal in $NdGa_6$ versus temperature.

3.4. $NdGa_6$

μSR measurements were performed at ISIS and at PSI. Two Lorentzian signals were observed, one fast and one slow. The ratio between the amplitudes was $A(\text{fast})/A(\text{slow}) \approx 3$.

The PSI data were obtained in zero field but, as the background signal was largely from copper, the damping of the slow component could not be extracted. The ISIS data were obtained in a 10 mT longitudinal field, sufficient to decouple the nuclear contribution to the depolarization. The two procedures gave the same result for the fast component. The results are presented in figures 5 and 6.

The depolarization of the fast component increases with a power law, $\lambda \sim (T - T_N)^{-0.61}$, where $T_N = 10.4$ K, for temperatures ranging from 300 K to 20 K. At 15 K, it was not possible to fit the data with a simple Lorentzian. This indicates that as the transition is approached, the depolarization becomes increasingly anisotropic, as observed in, for example, erbium [23]. Simulations are under way in order to investigate this possibility. It is likely that the magnetic clusters that remain in a temperature region above T_N reflect the spin structure found below T_N. Muons that enter a crystallite with the *c*-axis perpendicular to the polarization will then be much more strongly depolarized than those that enter with the polarization parallel.

As the magnetic structure of $NdGa_6$ is very simple, and as two distinct muon signals are obtained, this compound is a good candidate for studies of the influence of the dipole contribution and pair correlations on the depolarization. As seen in figure 6, the depolarization of the slow component saturates on approaching T_N, reflecting the fact that the average field for this site in the ordered state is zero.

The saturation occurs at about four times the ordering temperature, and a possible explanation is that it is at this temperature that clusters start to form.

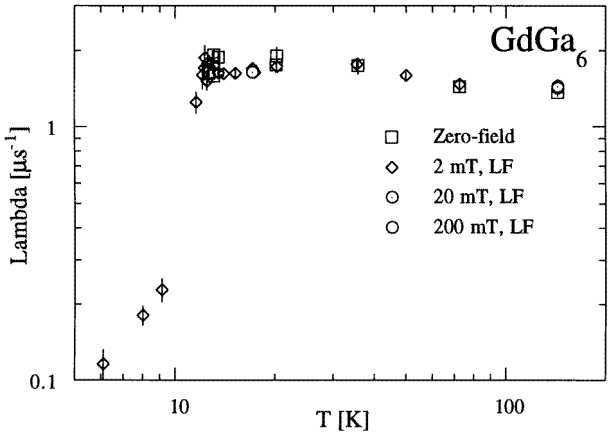

Figure 7. The temperature dependence of the exponential depolarization rate λ of the more rapidly decaying muon signal in $GdGa_6$.

3.5. GdGa$_6$

Two muon signals were observed in $GdGa_6$, one with a constant Lorentzian depolarization of $\lambda = 1.6(1)\ \mu s^{-1}$ between 144 K and 12.5 K and another with a Gaussian Kubo–Toyabe depolarization with $\Delta = 0.26(4)\ \mu s^{-1}$ also independent of temperature. In a 20 mT longitudinal field, the second component shows no depolarization, while the Lorentzian depolarization is not affected by longitudinal fields up to 200 mT. The ratio between the two amplitudes is *A*(Lorentzian)/*A*(Gaussian) ≈ 3.5. The amplitude and the damping of the

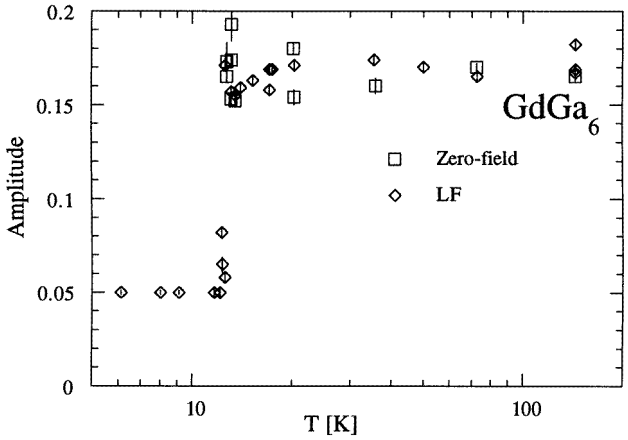

Figure 8. The amplitude of the more rapidly decaying exponential muon signal in $GdGa_6$ as a function of temperature.

fast component are displayed in figures 7 and 8.

At 12.5 K the amplitude of the fast component decreases to one third of its initial value as the compound orders magnetically. The amplitude of the slow component remains unchanged. This agrees with the proposed muon sites. The remainder of the fast component comes from the z-component of rapidly precessing muons and reflects the spin dynamics in the ordered state. The transition is very sharp, occurring over a temperature range of at most 0.1 K. Remarkable is the fact that no increase in depolarization rate is observed when the transition temperature is approached from above. Such an effect has been seen in single-crystal data—see, e.g., erbium metal [23]—if the muon spin is oriented parallel to the easy axis of magnetization, but is not expected for a polycrystal. However, recent data on single-crystalline gadolinium [24] also show little if any increase in damping rate close to T_c, independent of crystal orientation. A similar observation was made on $GdAl_2$ [12, 13]. This feature is probably due to the s-electron character of the Gd ion which makes it insensitive to crystal fields. The influence of the population of the CEF levels on the relaxation has been discussed by Noakes *et al* in 1987 [25]. Below the transition, the damping of the z-component decreases with temperature, which is normal in a magnetically ordered material.

The component that is only weakly damped in longitudinal fields shows Gaussian damping, caused by the nuclear magnetic dipoles, in zero external field both above and below the phase transition. This component must therefore come from muons at a zero-field site in the ordered state. However, given that the gadolinium moments fluctuate, there should also be a Lorentzian contribution to this signal. The statistics for the measurement were such that no accurate determination of the Lorentzian character can be made. As the Lorentzian depolarization of the other component is large, there must be a substantial difference between the distances to the nearest gadolinium atoms for the two sites assuming that the field fluctuation rates are identical. A possible site for the rapidly depolarizing muons is midway between two gadolinium atoms at a distance of 2.1 Å. For the muon site assumed, at $(0, 0, \frac{1}{2})$, the distance from the eight nearest gadolinium neighbours is 4.8 Å. By simple scaling, the Lorentzian depolarization for the $(0, 0, \frac{1}{2})$ site would then be about 0.04 μs^{-1} if the depolarization rate was 1.5 μs^{-1} at a site midway between the gadolinium

atoms. This is only slightly too high to be within the compounded systematic and statistical errors for the damping of the slow component in a 20 mT field.

The absence of any temperature-dependent behaviour for the depolarization indicates that pair correlations are very small. Since gadolinium has a spin-only magnetic moment, there are no crystal-field interactions that could stabilize the structure. It would be of interest to perform an x-ray diffraction study to investigate whether the transition involves any structural changes. Any magneto-elastic effects driving, for example, a first-order transition are, however, accompanied by hysteresis effects.

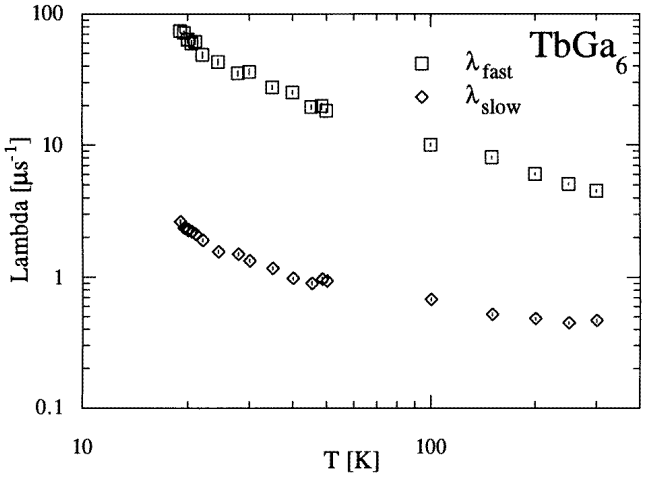

Figure 9. The muon depolarization above 20 K in $TbGa_6$ for the fast component (λ_{fast}) and above 19 K for the slow component (λ_{slow}).

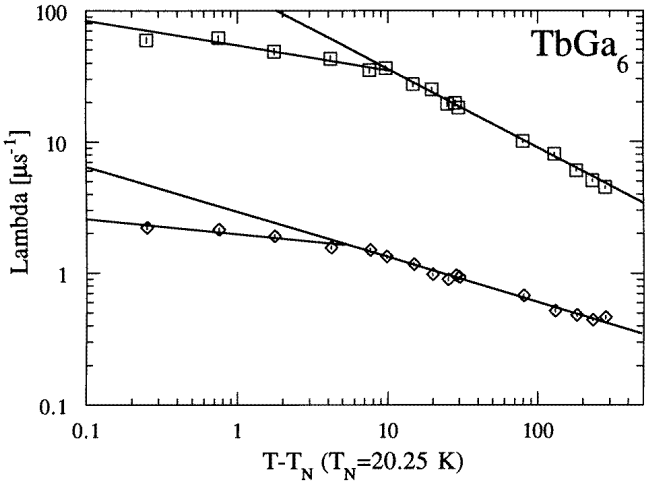

Figure 10. The depolarization of the two muon signals in $TbGa_6$ as a function of $T - T_N$ with $T_N = 20.25$ K.

3.6. $TbGa_6$

$TbGa_6$ was studied at TRIUMF in a helium-flow cryostat. Two muon signals were observed above the ordering temperature, one fast and one slow. The ratio between the amplitudes was $A(\text{fast})/A(\text{slow}) \approx 2.3$. T_{N1} was determined by the temperature at which the fast component would no longer fit the data with a constant amplitude. This was found to be between 20 and 20.5 K. As discussed above, the poor fits could also be due to an increase in the anisotropy of the depolarization. The value deduced for T_N from the change in the depolarization rate cannot be far from the true value given the size of the fitted moment at 18 K. The depolarization is plotted as a function of T in figure 9 and as a function of $T - T_{N1}$ in figure 10. The slow component could be followed into the incommensurately ordered state with certainty down to 17.5 K. Below this temperature it was no longer possible to differentiate between the fast and slow components. There was a slight shift in the base-line in the middle of the incommensurate region, but this had disappeared at 16 K.

The absolute value for the fast depolarization rate is strongly dependent on the value of the amplitude. The errors for the depolarization rate are therefore much larger than those shown in the figure, which are the statistical errors given by the least-squares fitting program.

As can be seen in figure 10, the depolarization does not diverge on approaching T_{N1} as a function of $T - T_{N1}$, but saturates instead. Down to about 10 K above the transition, the damping of the fast component follows a power law $\lambda \sim (T - T_N)^{-0.60(3)}$ while the slow component follows $\lambda \sim (T - T_N)^{-0.34(3)}$. The first value is identical within error to that obtained for $NdGa_6$, while the second one is strikingly similar to that obtained for $CeGa_6$. The slopes for the increase of the depolarization rate from 10 K above the transition temperature are $\lambda \sim (T - T_N)^{-0.18(3)}$ for the fast component and $\lambda \sim (T - T_N)^{-0.11(1)}$ for the slow one. It is therefore likely that short-lived magnetic clusters start to form at 10 K above the ordering temperature, changing the slope of the increase of the muon depolarization rate. Of course, it cannot be excluded that the change is due to a change in the population of crystal-field levels as discussed above in the section on $CeGa_6$. Unfortunately, the crystal-field levels of $TbGa_6$ are not known. It is possible that the crystal-field levels scale with the ordering temperature but that would be a coincidence. The formation of short-lived magnetic clusters is therefore, in the view of the authors, a more probable explanation.

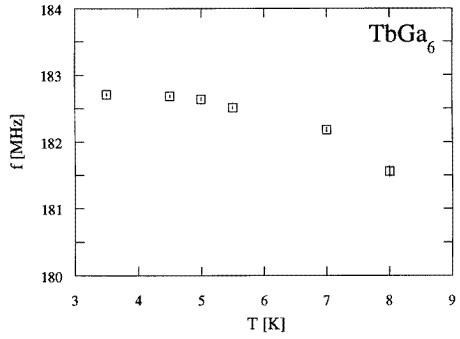

Figure 11. The precession frequency in $TbGa_6$ as a function of temperature.

It is also interesting to observe that the difference between the exponents of the two power laws for $TbGa_6$ depends solely on the muon site. The saturation on approaching the ordering temperature in $TbGa_6$ is likely to stem partly from the nature of the ordering and partly from the details of the dipole tensor for different $\boldsymbol{k}$-vectors for the given site. The

saturation of the depolarization for the slow component is not unexpected given that the static moment is zero in the ordered state.

It would be interesting to perform additional measurements on $NdGa_6$ in the high-temperature region with improved statistics, to see whether the slow component increases with a power law similar to that observed for $CeGa_6$ and $TbGa_6$.

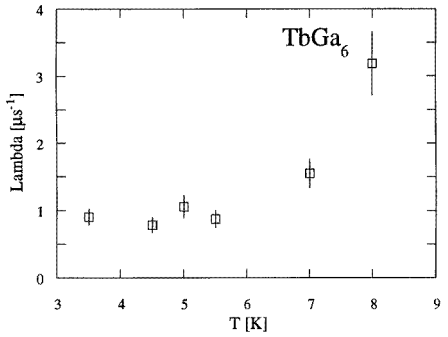

Figure 12. The damping of the precessing signal in $TbGa_6$ as a function of the temperature.

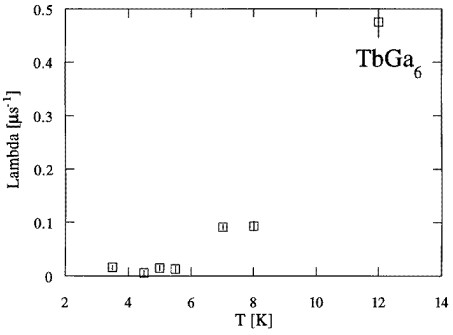

Figure 13. Damping of the one third of the polarization vector for the precessing muons that is parallel to the z-direction plotted as a function of temperature.

Below T_{N2}, three components were found. The first was a precessing component with a frequency of 182 MHz; see figure 11. The amplitude was only between 0.022 and 0.025, but the time bins used in the experiment require the amplitude to be multiplied by a factor of three to four to take time averaging into account. The damping of the precessing signal is plotted in figure 12. This signal shows thus the typical behaviour for a magnetically ordered phase. There was also a very weakly damped component that had an amplitude of about half that of the corrected amplitude of the precessing signal and this is the third of the initial polarization that was parallel to the z-axis. The damping of this signal is plotted in figure 13. Thirdly, there was a component with a rather large depolarization. This component was interpreted as coming from muons stopping at the zero-field sites, i.e. the site that gives the muon signal with the lower depolarization rate in the paramagnetic region.

4. Conclusions

It is shown that the $REGa_6$ series presents an example of very varied magnetic ordering behaviour within compounds that are structurally very similar. In $CeGa_6$, $NdGa_6$ and $TbGa_6$ a systematic build-up of magnetic correlations occurs, while in $GdGa_6$ no increase in correlations is observed within the experimental resolution of 0.1 K. The different ordering characteristics for the gadolinium compound might indicate the importance of the population of the CEF levels in the other compounds. In the case of $NdGa_6$ the correlations have been observed at temperatures 30 times the ordering temperature. Given the anisotropy of the structure, it is likely that correlations are much stronger in the rare-earth planes than between them. It could then be speculated that correlations first build up within planes, but that the structures do not order until the temperature is low enough for the interactions between the planes to create three-dimensional ordering. We do not understand the sudden ordering of the gadolinium compound. An investigation should be performed to investigate whether

the transition involves any structural changes. In all of the compounds, except $CeGa_6$, two distinct muon sites are observed. The exponents for the damping as a function of the reduced temperature are critically dependent on the muon site. The likely explanation is a substantial difference between the temperature dependences of the relative weights of the dipolar tensor for different $\boldsymbol{k}$-vectors. This is particularly well demonstrated in $TbGa_6$ where the depolarization for the two different sites was most easily followed.

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